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(54) Electrochemical Gas Generator

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ELECTROCHEMICAL GAS GENERATOR

Field of the Invention

The present invention relates to a device for electrochemical generation of gases for the transportation of fluids, lubricants and similar media.

Background of the Invention

It is known to use catalytical and electrochemical processes to produce pressurized gases for transporting fluids in technical applications. For example, hydrazine can be catalytically decomposed into hydrogen and nitrogen thereby generating a pressurized gas which can be used to quickly discharge the water filled tanks of submarines. The gas mixture produced by this reaction contains ammonia in a concentration which is a function of the nature of the catalyst. The higher the proportion of ammonia the higher the temperature of the generated gas. The pressurized gas may be used, for example, in control units in space applications.

It is also possible to use oxygen as a pressurizing gas. Oxygen gas may be generated by catalytically decomposing hydrogen peroxide using, for example, a silver catalyst. This reaction also produces a large quantity of heat which, in general, requires special heat management.

With both of the methods described above, the rate at which gas is released is determined by the diffusion or convection current of the reaction fluid to the catalyst. Therefore the reaction may only be stopped by interrupting the current.

It has been proposed to use self-controlled decomposition apparatuses for hydrazine and hydrogen peroxide by using catalysts similar to those used in the tech-

nique of gas diffusion electrodes (valve electrodes are disclosed in US patent 3 201 282 and German patent 1 542 565). In these systems the rate of reaction is dependent upon the pressure of the generated gas. By using a diluted aqueous solution of hydrazine (or hydrogen peroxide) the rate of gas production may be kept constant provided that the discharge of water is kept constant.

It is also possible to generate hydrogen gas by reaction of a metal with a base or an acid. When hydrochloric acid is brought into contact with zinc, hydrogen is generated, and zinc chloride is released into solution. Hydrogen can also be generated by the reaction of zinc in an alkaline solution but when using pure zinc almost no generation of hydrogen can be observed. This is because the hydrogen minimum overvoltage of zinc is extremely high. The inertness of zinc in an alkaline medium is also due, in part, to the generation of a layer of zinc oxide on the surface of the zinc which passivates the zinc.

It is possible to accelerate the corrosion by contaminating the zinc with another metal having a much lower hydrogen minimum overvoltage. For example, it is well known that when a clean zinc plate is contacted by a platinum wire Hydrogen gas is generated near the wire and the zinc corrodes close to the contact area.

It is also known to drill a hole in a zinc block and to solder the block onto a Molybdenum bar. Again, the result is a galvanic element. The rate of generation of hydrogen depends upon the size of the metal surface upon which the generation of Hydrogen is promoted. The performance of a short-circuit element such as this depends upon many random factors relating to how the surfaces influencing the corrosion are formed. This means that it is not easily possible to regulate the rate of generation of hydrogen from outside.

Lubricant dispensers which dispense lubricant by means of a gas produced by a galvanic element and an electrolytic fluid are well known. To start the generation of gas, the galvanic element is brought into contact with the electrolyte by means of a screw. The generated gas expands a hollow body and moves a piston or a dividing insert which presses the lubricant out of a grease box to the lubrication point (German patent 2 139 771 corresp. to CA 961 420).

Summary of the Invention

The present invention pertains to a device for generating gases which device is able to produce gases in an adjustable quantity and/or at exact defined times, which is compact, easy to manufacture and which prevents the penetration of electrolyte.

In particular the invention pertains to an electrochemical gas generator comprising a galvanic cell and a means for completing an electrical circuit between the electrodes of the cell. The galvanic cell comprises a sealed housing, a gas generation electrode within the housing, a counter electrode within the housing but separated from the gas generation electrode, an electrolyte in contact with the gas generation electrode and the counter electrode, an aperture in the housing to release gas generated at the gas generation electrode, a path by which gas can flow from the gas generation electrode to the aperture and a gas permeable, electrolyte impermeable, membrane blocking the path between the gas generator electrode and the aperture.

Another embodiment of the device according to the invention comprises a galvanic cell including an anode, a cathode and a housing containing an aqueous electrolyte in

which by closing of an external circuit, and, if necessary by supplying a source of direct current in the external circuit, a current flows which produces a quantity of gas corresponding to the current.

The invention is directed to a device for electrochemically generating a hydrogen gas or an oxygen gas in an adjustable quantity, comprising: a galvanic cell including an enclosed housing having a base and at least one side wall integral with said base, a cover, a seal positioned between and in contact with said cover and said side wall of said housing, and an opening for releasing gas from said housing, said galvanic cell further including; a gas generating electrode forming a part of said housing and including an electron-conducting porous body contained within said housing; a counter electrode including an oxidizable metal or a reducible oxide or nitrate which serves in countercapacity to said gas generating electrode; separator means between said gas generating electrode and said counter electrode; an alkaline electrolyte present in an amount sufficient to provide reactions in said gas generating electrode and in said counter electrode; and means for electrically connecting said gas generating electrode and said counter electrode, and means for establishing a current flow between said gas generating electrode and said counter electrode for generating a predetermined quantity of gas by said gas generating electrode, for release from said opening of said galvanic cell.

The invention is also directed to an actuating apparatus for means for dispensing solids and fluids, or transporting mediums, including the device defined, which also possesses means connecting said opening of said galvanic cell and said dispensing means or said transporting medium, for supplying gas to said actuating apparatus for operating said dispensing means or said transporting medium.

The device as defined for generating hydrogen gas in an adjustable quantity wherein said electron-conducting porous body of said gas generating electrode forming a part of said housing may include a hydrophilic, electrolyte-receiving portion and a hydrophobic, gas-receiving portion, and a porous hydrophobic layer adjacent to and in engagement with said porous body, for preventing electrolyte from leaving the galvanic cell; and wherein said counter electrode may include an oxidizable metal which serves in countercapacity to said gas generating electrode; and may include means for excluding air from said galvanic cell, and for excluding oxygen from the pores of the gas generating electrode.

The device as defined for generating oxygen gas in an adjustable quantity wherein said electron-conducting porous body of said gas generating electrode forming a part of said housing may include a hydrophilic, electrolyte-receiving portion and a hydrophobic, gas-receiving portion, and a porous hydrophobic layer adjacent to and in engagement with said porous body, for preventing electrolyte from leaving the galvanic cell; and said counter electrode may include a reducible oxide which serves in countercapacity to said gas generating electrode; and means for electrically connecting said gas generating electrode and said counter electrode, and for establishing a current flow between said gas generating electrode and said counter electrode for generating a predetermined quantity of gas by said gas generating electrode, for release from said opening of said galvanic cell.

The invention is also directed to a method for electrochemically generating a hydrogen gas or an oxygen gas in an adjustable quantity by means of a galvanic cell including an enclosed housing having a base and at least one side wall integral with said base, a cover, a seal

positioned between and in contact with said cover and said side wall of said housing, and an opening for releasing gas from said housing, said galvanic cell further including a gas generating electrode forming a part of said housing and including an electron-conducting body contained within said housing, a counter electrode including an oxidizable metal or a reducible oxide which serves in countercapacity to the gas generating electrode, separator means between said gas generating electrode and said counter electrode, and an alkaline electrolyte present in an amount sufficient to provide reactions in said gas generating electrode and in said counter electrode, said method comprising the steps of: electrically connecting said gas generating electrode and said counter electrode, establishing a current flow between said gas generating electrode and said counter electrode, generating a predetermined quantity of gas by said gas generating electrode, and releasing said gas from said opening of said galvanic cell.

In the device as defined, said oxidizable metal can be selected from the group consisting of zinc, cadmium, lead and copper. Said reducible oxide can be selected from the group consisting of manganese dioxide, silver oxide, mercury oxide and nickel oxide. Said reducible oxide can be selected from the group consisting of nitrate or ammonium nitrate.

Said gas generating electrode can comprise Raney-metals of Group VIII of the periodic table or noble metals selected from the group consisting of Pt, Pd and platinum containing metals. Said porous body of said gas generating electrode can comprise a metal from Group VIII of the Periodic Table of elements selected from the group consisting of platinum, palladium and nickel, alone or in combination with carbon, said metals having low overvoltage with respect to the evolution of the gas at said electrode.

Said gas generating electrode can generate hydrogen gas and can comprise a Raney nickel powder bound with a porous foil of polytetrafluoroethylene metal rolled into a net of nickel, said rolled nickel net being a current conductor. Charcoal powder can be blended with said Raney nickel powder.

Said gas generating electrode can generate oxygen gas and can comprise Raney nickel powder in admixture with a hydrophobic resin powder selected from the group consisting of polytetrafluoroethylene and polyethylene. Said oxidizable metal can be zinc powder or zinc gel and said cell can operate at a voltage of about 0.4V or less. Said zinc cell can be short circuited via low resistance and can contain sealing means to prevent entrance of air into said cell when said cell starts generating gas to actuate said means for dispensing or transporting.

Said opening in said housing can release generated gas resulting from electrochemical activity within said cell and air can be excluded from said device by valves which allow a one directional flow of the generated gas to actuate. An electrical circuit and adjustable resistance can be provided to control gas generation.

The device may contain means to open the circuit provided by direct current upon reaching a predetermined gas pressure and to close said circuit when said gas pressure falls below a predetermined value. The generated gas can be released in pulses.

Said oxidizable metal can be zinc gel containing said electrolyte situated within said housing. The said reducible oxide can be in the form of a porous tablet of a manganese dioxide and can be situated within said housing.

Brief Description of the Drawings

Figure 1 is a cross section through a cell made according to the invention.

Detailed Description of the Preferred Embodiment

It has been found that it is advantageous to produce hydrogen in a cell having a cathode made of zinc powder and an anode, which is a hydrogen evolution electrode, in the form of a gas diffusion electrode. In a preferred embodiment the gas evolution electrode is made of a metal of the 8th sub group of the periodic table of elements, preferably platinum, palladium or nickel, the latter in particular in the form of Raney nickel, because these metals have a low hydrogen minimum overvoltage. The electrode is preferably made in form of a porous electrode, e.g. as a double-skeleton catalyst electrode according to German patent 1 019 361 or in form of a Raney-nickel structure combined with PTFE (polytetrafluorethylene) or in the form of a Raney-nickel/active charcoal-structure embedded in the meshes of a net or of an expanded metal. The cell containing the zinc, electrolyte, a separator and the hydrogen evolution cathode is closed to the outside by a foil of PTFE. The PTFE foil allows gas to pass through its pores. At the same time the high capillary depression in the pores of the unwettable PTFE prevents the exit of electrolyte from the cell.

It has been found that zinc/air cells which are typically used in hearing aids can be used - against their proper purpose - for the new object of generating hydrogen by making a short-circuit via a low resistance and sealing the cell against entrance of air in the starting phase. Thus, evolution of hydrogen and flow of a current start. The evolution of hydrogen is maximized where the electromotive force of the potential difference between the

reversible zinc electrode and the reversible hydrogen evolution electrode, is about 0.5 volts. By selection of the proper short-circuit resistance it is possible to calculate the corrosion current and to regulate the rate at which hydrogen is produced by temporarily changing the incremental resistance of the galvanic cell.

The cathode normally used in such known zinc/air cells is not applicable to the new purpose. It is possible to adapt this element in an optimal way to the new purpose by exchanging the electrode made of a compound of PTFE and active charcoal for another made of a compound of Raney-nickel and PTFE or Raney-nickel and charcoal which compound is laminated or rolled into a net of nickel and by providing the layer with a porous foil of PTFE according to German patent application 3 342 969. In principle it is possible to use a massive zinc cup, zinc powder or a gel of zinc powder, which is normally used in production of primary cells in the battery industry, as the zinc electrode. In order to reduce the unwanted evolution of hydrogen by self-corrosion the zinc may be amalgamated.

For discharging the zinc powder or zinc gel electrode is in contact with an amalgamated metal nail or a galvanized or cadmium-protected contact element of relatively high hydrogen minimum overvoltage. This can also be a part of the housing of the cell made of a corresponding metal, such as zinc or brass. This housing part is separated by an electrically isolating seal from a second metallic housing part which includes the gas evolution electrode. The quantity of zinc which is required in the housing depends upon the quantity of hydrogen to be produced in the cell. By contrast, the size of the gas evolution electrode depends only upon the maximum rate of hydrogen evolution required. For a quantity of 40 Ncm^3/h of hydrogen, corresponding to 100 mamps an electrode having a surface area of 1 cm^2 is sufficient.

The quantity of electrolyte required is proportional to the quantity of zinc. During the reaction water is consumed, while hydrogen and zinc oxide are produced as reaction products. The quantity of electrolyte must be such that after the consumption of the water needed for the reaction enough electrolyte fluid remains.

In the zinc/air cell oxygen from the air is absorbed, therefore the volume of the cell increases as gas generation proceeds. A zinc/air cell must be dimensioned according to the final volume of the balance of the produced and consumed materials. The same relates to the hydrogen producing cell but with the difference that the hydrogen produced in high volume needs only little space within the cell. During reaction a reduction of total volume takes place because the hydrogen produced also transports vapour out of the housing. To maintain contact between the phases involved in the reaction, the electrolyte may be pressurized, which can be done by introducing it with small overpressure into the wide pores of a hydrophobe body or by using hydrophile absorbent paper in the important areas. It is also possible to maintain contact between a zinc electrode and conductor and separator by means of a spring element, the separator is a hydrophile and arranged between the zinc electrode and the gas evolution electrode.

Finally, it is possible to enforce the change of volume from outside by deformation of the cell housing.

The element disclosed may be applied to pressure pistons used to transport fluids and similar media. Due to the fact that the internal resistance of such cells is only some ohms a slow rate of gas generation may be set and controlled by varying the external resistance. As electromotive force the difference in voltage between the zinc

electrode and the reversible hydrogen electrode may be regarded which in 6 n (normal) potash lye (KOH) is 0.42 volts. To produce a hydrogen current which is equivalent to an electric current of 10 mamps an external resistance of 50 ohms is sufficient. To produce hydrogen equivalent to an electric current of 1 mamps the external resistance must be 500 ohms. In this case the variation of the internal resistance of the cell of about 10 ohms is of no importance any longer.

The gas generating element according to the invention is arranged within the space of the piston or in the pressure chamber. It will be activated e.g. by a sealed screw which via a chosen resistance produces a short-circuit between the zinc electrode and the gas producing electrode. Preferably the housing of the cell may be formed by deformation of the piston while the gas evolution electrode is arranged within the housing in a sealed and pressure-safe manner. To start gas generation, a short-circuit is made between the zinc electrode and the gas evolution electrode by a resistance which e.g. is formed as a layer and comprises a time-scale for adjusting the process time, which scale shows in which time the available quantity of lubricant will be dispensed.

In the example mentioned hydrogen will be produced because the whole active volume of the cell is filled with a substance which will be oxidized by water. If the zinc is exchanged for cadmium it is not possible to start hydrogen generation by contact of a metal with a low hydrogen minimum overvoltage. However, such an element may be used for hydrogen evolution by running a current through the element from an external source and arranging the cadmium as anode and the hydrogen evolution electrode as cathode. In this case the cathode is connected to the negative pole and the cadmium electrode is connected to the positive pole of the electric source. Also in this cell

hydrogen is produced which is equivalent to the current. In contrast to the zinc element described above there is no danger of hydrogen evolution in the absence of current, because cadmium is a noble metal with respect to hydrogen and is not able to displace hydrogen out of the compound with oxygen.

In some cases it is preferable to block evolution of hydrogen. In such cases an oxygen producing element may be used in which oxygen is anodically produced in the electrodes in the described manner. As a counter electrode a metal oxide is used, e.g. silver oxide, mercury oxide, nickel oxide, lead dioxide or manganese dioxide; in such cases the oxides are reduced - depending on their electrochemical behaviour - to metal (silver, mercury etc.) or to an oxide with lower valency (e.g. Mn_3O_4). However, the current consumption to produce a corresponding quantity of oxygen is twice that required for generating hydrogen because the electrochemical valency of a molecule of oxygen is 4 while the electrochemical valency of hydrogen is only 2.

There is no need to use metal oxides as material for oxygen electrodes. It is possible to reduce ions of nitrate into ammonia in a special cell, while the counter electrode is producing oxygen. Raney-nickel electrodes may be used as both oxygen and nitrate reduction electrodes. It is also possible to use ammonia for generation of hydrogen because under anodic current the ions of ammonia are oxidized to nitrite resp. nitrate ions in the Raney-nickel electrodes, while the counter electrode produces hydrogen.

The common features of the gas producing cells described are, that they contain the initial state either:

a) electrochemical oxidizable substances, a hydrogen electrode and a liquid electrolyte or

b) electrochemical reducible substances, an oxygen electrode and a liquid electrolyte

and that they generate under flow of current initiated from outside hydrogen or oxygen, which is produced in the pores of a gas diffusion electrode and penetrates through the pores of a hydrophobe diffusion membrane into an external chamber, while the electrolyte is retained in the cell housing due to the high capillary depression of the membrane.

Detailed Description of the Drawing

Figure 1 shows a diagrammatic elevation in section of one embodiment of a cell according to the invention. The knob cell comprises a cup 1 and a cover 2 which together with a plastic seal 3 form the housing. Within the cover 2 and in contact with it is an active substance 4 in form of a zinc gel containing an electrolyte or in form of a porous tablet of manganese dioxide. 5 is a compressible porous body which may contain an additional quantity of electrolyte. 6 is a fleece impregnated with electrolyte, 7 a separator in form of a ion-exchange foil. This foil is kept in position by a support ring 8. 9 is the gas diffusion electrode which is e.g. made of a Raney-nickel powder bound with PTFE and rolled into a net of nickel. On the side to the bottom of the cup 1 the gas diffusion electrode is provided with a PTFE foil. The metallic support ring 8 is in contact with the gas diffusion electrode 9 and electrically connects the gas diffusion electrode 9 and the cup 1. 10 is a wide-pore fleece layer which channels the gas generated in the gas diffusion

electrode to the hole 11 in the bottom of the cup from where is leaves the cell.

Every zinc atom sets free two electrons, which are able to reduce one molecule of water to hydrogen. It is therefore necessary to insert into the cell for every 65 grams of zinc 18 grams of water.

As will be apparent to those skilled in the art in the light of the foregoing disclosure, many alterations and modifications are possible in the practice of this invention without departing from the spirit or scope thereof. Accordingly, the scope of the invention is to be construed in accordance with the substance defined by the following claims.

WHAT IS CLAIMED IS:

1. A device for electrochemically generating a hydrogen gas or an oxygen gas in an adjustable quantity, comprising:

a galvanic cell including an enclosed housing having a base and at least one side wall integral with said base, a cover, a seal positioned between and in contact with said cover and said side wall of said housing, and an opening for releasing gas from said housing, said galvanic cell further including;

a gas generating electrode forming a part of said housing and including an electron-conducting porous body contained within said housing;

a counter electrode including an oxidizable metal or a reducible oxide or nitrate which serves in counter-capacity to said gas generating electrode;

separator means between said gas generating electrode and said counter electrode;

an alkaline electrolyte present in an amount sufficient to provide reactions in said gas generating electrode and in said counter electrode; and

means for electrically connecting said gas generating electrode and said counter electrode, and means for establishing a current flow between said gas generating electrode and said counter electrode for generating a predetermined quantity of gas by said gas generating electrode, for release from said opening of said galvanic cell.

2. An actuating apparatus for means for dispensing solids and fluids, or transporting mediums, including the device defined in claim 1, which also possesses means connecting said opening of said galvanic cell and said dispensing means or said transporting medium, for supplying gas to said actuating apparatus for operating said dispensing means or said transporting medium.

3. The device defined in claim 1 for generating hydrogen gas in an adjustable quantity wherein said electron-conducting porous body of said gas generating electrode forming a part of said housing includes a hydrophilic, electrolyte-receiving portion and a hydrophobic, gas-receiving portion, and a porous hydrophobic layer adjacent to and in engagement with said porous body, for preventing electrolyte from leaving the galvanic cell;

and wherein said counter electrode includes an oxidizable metal which serves in countercapacity to said gas generating electrode;

and includes means for excluding air from said galvanic cell, and for excluding oxygen from the pores of the gas generating electrode.

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4. The device defined in claim 1 for generating oxygen gas in an adjustable quantity wherein said electron-conducting porous body of said gas generating electrode forming a part of said housing includes a hydrophilic, electrolyte-receiving portion and a hydrophobic, gas-receiving portion, and a porous hydrophobic layer adjacent to and in engagement with said porous body, for preventing electrolyte from leaving the galvanic cell;

and said counter electrode includes a reducible oxide which serves in countercapacity to said gas generating electrode; and

means for electrically connecting said gas generating electrode and said counter electrode, and for establishing a current flow between said gas generating electrode and said counter electrode for generating a predetermined quantity of gas by said gas generating electrode, for release from said opening of said galvanic cell.

5. A method for electrochemically generating a hydrogen gas or an oxygen gas in an adjustable quantity by

means of a galvanic cell including an enclosed housing having a base and at least one side wall integral with said base, a cover, a seal positioned between and in contact with said cover and said side wall of said housing, and an opening for releasing gas from said housing, said galvanic cell further including a gas generating electrode forming a part of said housing and including an electron-conducting body contained within said housing, a counter electrode including an oxidizable metal or a reducible oxide which serves in countercapacity to the gas generating electrode, separator means between said gas generating electrode and said counter electrode, and an alkaline electrolyte present in an amount sufficient to provide reactions in said gas generating electrode and in said counter electrode, said method comprising the steps of:

electrically connecting said gas generating electrode and said counter electrode, establishing a current flow between said gas generating electrode and said counter electrode, generating a predetermined quantity of gas by said gas generating electrode, and releasing said gas from said opening of said galvanic cell.

6. The device defined in claim 3 wherein said oxidizable metal is selected from the group consisting of zinc, cadmium, lead and copper.

7. The device defined in claim 4 wherein said reducible oxide is selected from the group consisting of manganese dioxide, silver oxide, mercury oxide and nickel oxide.

8. The device defined in claim 4 wherein said reducible oxide is selected from the group consisting of ammonia and ammonium nitrate.

9. The device defined in claim 1 wherein said gas generating electrode comprises Raney-metals of Group VIII

of he periodic table or noble metals selected from the group consisting of Pt, Pd and platinum containing metals.

10. The device of claim 3 wherein said porous body of said gas generating electrode comprises a metal from Group VIII of the Periodic Table of elements selected from the group consisting of platinum, palladium and nickel, alone or in combination with carbon, said metals having low overvoltage with respect to the evolution of the gas at said electrode.

11. The device defined in claim 9 wherein said gas generating electrode generates hydrogen gas and comprises a Raney nickel powder bound with a porous foil of polytetrafluoroethylene metal rolled into a net of nickel, said rolled nickel net being a current conductor.

12. The device defined in claim 8 wherein charcoal powder is blended with a Raney nickel powder.

13. The device defined in claim 8 wherein said gas generating electrode generates oxygen gas and comprises Raney nickel powder in admixture with a hydrophobic resin powder selected from the group consisting of polytetrafluoroethylene and polyethylene.

14. The device defined in claim 6 wherein said oxidizable metal is zinc powder or zinc gel and said cell operates at a voltage of about 0.4V or less.

15. The device defined in claim 14 wherein said zinc cell is short circuited via low resistance and contains sealing means to prevent entrance of air into said cell when said cell starts generating gas to actuate a means for dispensing solids and fluids, or transporting mediums.

16. The device defined in claim 1 wherein said opening in said housing releases generated gas resulting from electrochemical activity within said cell and air is excluded from said device by valves which allow a one directional flow of the generated gas to actuate.

17. The device defined in claim 2 wherein an electrical circuit and adjustable resistance is provided to control gas generation.

18. The device defined in claim 2 containing means to open a circuit provided by direct current upon reaching a predetermined gas pressure and to close said circuit when said gas pressure falls below a predetermined value.

19. The device defined in claim 2 wherein the generated gas is released in pulses.

20. The device defined in claim 14 wherein said oxidizable metal is a zir el containing said electrolyte and is situated within said housing.

21. The device in claim 7 wherein the said reducible oxide is in the form of a porous tablet of a manganese dioxide and is situated within said housing.

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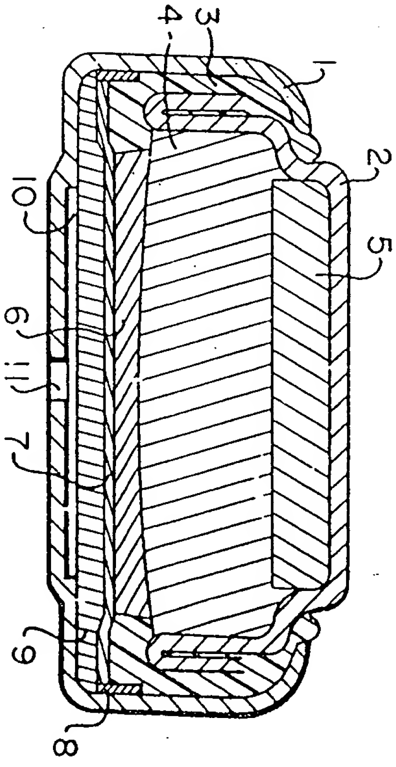

FIGURE 1